



## Understanding defect related luminescence processes in wide bandgap materials using low temperature multi-spectroscopic techniques

Prasad, Amit Kumar

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in the Basongcuo Catchment, Eastern Nyainqentanglha are consistent with those in the surrounding area. Comparing the ages of glacier advances with the insolation, effective moisture and speleothem records from Dongge-Hulu cave, we argue that the glacier advances in the Basongcuo Catchment were also controlled by temperature.

Those who are interested in this thesis can ask the Dr Gang Hu (hugang@itpcas.ac.cn) or Dr. Chaolu Yi (clyi@itpcas.ac.cn) for provision.

**Amit Kumar Prasad**

**Understanding defect related luminescence processes in wide bandgap materials using low temperature multi-spectroscopic techniques**

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Center for Nuclear Technologies, Technical University of Denmark, Risø Campus, Roskilde 4000

*Degree: Ph.D.*

*Supervisors: Dr. Mayank Jain (main supervisor) and Dr. Torben Lapp (co-supervisor)*

Feldspar is a dominant, naturally occurring mineral that comprises about ~60 % of the Earth's crust. It is widely used in optically stimulated luminescence (OSL) dating of sediments to obtain chronologies of past events as old as ~0.5 Ma, and thus, plays a crucial role in understanding Quaternary climate changes, landscape development and human evolution and dispersal. Optical properties of feldspar originate from a) a wide band gap (~7.7 eV), b) crystal defects (impurity atoms and distortions) that create localized energy states within the bandgap, and c) conduction band and the low-mobility band tail states, which play a role in charge transport. Despite a rapid progress in the infra-red stimulated luminescence (IRSL) dating technique using feldspar, a clear understanding of luminescence process is still lacking. A better understanding of feldspar as a physical system is expected to lead to its improved exploitation as a luminescence chronometer. My Ph.D. investigates the nature of luminescence generating defects and processes in feldspar, and tests whether the intra-defect relaxation transitions may be successfully used to improve the dating technique. It includes mapping the energy states of defects individually and characterizing their emission process, understanding the dynamics of the excited-state relaxation and tunneling, and defect interactions with the crystal lattice and the band tail states. The experiments were carried out using the Risø station for Cryogenic Luminescence Research (COLUR) and a high sensitive spectrometer attached to the Risø TL/OSL reader. The key findings of my Ph.D. research are summarized as follows:

1) I discovered the excitation-energy dependent emission (a red edge effect) in the green-orange emission in feldspar, and demonstrated that this effect arises from interaction of a deep lying defect with the band tail states. This effect can be used to measure the band-tail width through relatively simple spectroscopic (photoluminescence) measurements.

2) My studies on  $\text{Fe}^{3+}$  show that its deep red emission varies with site dependence of  $\text{Fe}^{3+}$  even within a single sample. Furthermore, it is observed that there exists an excitation-energy dependence of the main radiative transition ( ${}^4\text{T}_1 \rightarrow {}^6\text{A}_1$ ) in  $\text{Fe}^{3+}$ ; this is possibly related to spin-lattice interaction.

3) I explored a model analogue system for feldspar called  $\text{YPO}_4\text{:Ce,Sm}$ , in order to understand IRSL produced by excited-state tunneling. For the first time, a precise mapping of the energy levels of the metastable  $\text{Sm}^{2+}$  was carried out, and the temperature-dependent relaxation lifetime of  $\text{Sm}^{2+}$  excited state was determined using the defects internal radiative-transition. It was then demonstrated that OSL decay curves resulting from optically induced, sub-conduction band electron transfer ( $\text{Sm}^{2+} \rightarrow \text{Ce}^{4+}$ ) can be adequately described using the prevalent mathematical model of excited-state tunneling.

4) Finally, inspired by the results of  $\text{YPO}_4\text{:Ce,Sm}$ , I discovered a Stokes-shifted, infra-red photoluminescence (IRPL) signal arising from the principal trap in feldspar (excitation ~ 1.4 eV (885 nm), emission: ~ 1.3 eV (950 nm)). Current methods of OSL rely on transfer of electrons from the principal trap to holes located elsewhere in the lattice; this is by default a destructive readout of dosimetric information. Furthermore, OSL (or IRSL) suffer from sensitivity changes because of competition in the recombination process, leading to possible uncertainties in the dose measurement. In contrast to IRSL, the IRPL signal arises from intra-defect excitation and the subsequent radiative relaxation within the principle trap (i.e. the trap giving rise to IRSL). IRPL is a non-destructive readout technique and the lifetime of the excited state relaxation is estimated to be ~40  $\mu\text{s}$  at 7K and ~29  $\mu\text{s}$  at 295 K. The IRPL signal increases with dose and the preliminary dating investigations indicate that this signal contains an athermal non-fading component, likely arising from the trapped electrons that do not have a nearby hole center.

There are two important technique developments in my thesis. Firstly, based on the model of the red edge effect, a simple method is proposed for estimation of the width of the band tail states in feldspar. Secondly, it is shown that the new IRPL signal can be used for non-destructive probing of dosimetric information in the principal trap. The IRPL technique is likely to provide a) a robust understanding of the behavior of electron trapping centers in feldspar, b) a possibility of selective probe of non-fading electrons without using any thermal assistance, and c) precise measurements of luminescence from very small volumes by repeated readout. These possibilities open new windows for development of robust dating methods as well as advanced imaging techniques. I envision that the IRPL signal will significantly impact the field of optical dating.

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